Determination of the border between "shallow" and "deep" tunneling regions for Herman-Kluk method by asymptotic approach

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Abstract

The evaluation of a tunneling tail by the Herman-Kluk method, which is a quasiclassical way to compute quantum dynamics, is examined by asymptotic analysis. In the shallower part of the tail, as well as in the classically allowed region, it is shown that the leading terms of semiclassical evaluations of quantum theory and the Herman-Kluk formula agree, which is known as an *asymptotic equivalence*. In the deeper part, it is shown that the asymptotic equivalence breaks down, due to the emergence of unusual "tunneling trajectory", which is an artifact of the Herman-Kluk method.

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Even nowadays, it is still impossible to carry out serious numerical investigation of quantum dynamics, even for modest (e.g., 10–100) degrees of freedom systems, with the present state of the art of computational technology, unless we take drastic approximations that need to be based on good physical insights. As a starting point to invent such a method, it is often employed the semiclassical approximation (i.e. asymptotic evaluation) of the path integral representation of time evolution operator (Feynman kernel) [1]. The semiclassical approximation, however, has difficulties due to the exponential proliferation of contributing trajectories and caustics (see, e.g. Ref. [2]) and due to Stokes phenomenon that requires to remove non-contributing complex-valued trajectories [3], even in few degrees of freedom systems, when the corresponding classical system is chaotic. Furthermore, in the semiclassical method, the boundary conditions of the contributing classical trajectories both for initial and finial times, is troublesome (known as a root-search problem [4, 5]), in computations for realistic systems such as atoms and molecules. In order to avoid the root-search problem, initial value representations (IVRs) of Feynman kernels are proposed. The IVRs imposes only the initial conditions on the classical trajectories. In Ref. [4], the earliest version of IVR is introduced by a change of integral variables to semiclassical Feynman kernel. A general framework of IVR is proposed by Kay [6], who discussed various integral expressions (IEs) of approximate Feynman kernel, which are composed by classical trajectories that are emitted from real-valued initial conditions. The important guiding principle of Kay's theory is that the leading semiclassical expressions of both an IE and exact quantum theory must agree. This is called *the asymptotic equivalence* [6, 7]. Furthermore, Kay argued that several known IVRs (IEs), including thawed Gaussian approximation [8], cellular dynamics [9], and the Herman-Kluk (HK) formula [10], are asymptotically equivalent with quantum theory. Nowadays, a lot of numerical investigations of quantum dynamics, including rather realistic systems, employ IVRs, in particular, the Herman-Kluk method [11].

The limitation of IVRs, however, is not clear [12], in particular, in the descriptions of classically forbidden processes, e.g., tunneling processes, whose conventional semiclassical treatments need to take into account the contributions from complex-valued classical trajectories. Numerical experiments to reproduce tunneling tails by IVR approaches suggest that the "shallow" side of tunneling tails is tractable [13]. On the other hand, concerning to the "deep" side, the IVR approaches have difficulties. Kay's *semiclassical* analysis of $\mathcal{O}(\hbar^2)$ error term of Herman-Kluk method reveals that the magnitude of the error is controlled

by the *complex-valued* classical trajectories [14]. However, these works are not conclusive. First, since the tunneling tails are exponentially small, the corresponding error analysis also requires to treat *exponentially small errors*. Hence, the $\mathcal{O}(\hbar^2)$ error term, which is satisfactory in classically allowed region, is too large. Second, the border between the shallow side and the deep side of the tunneling tail has been unknown. In order to clarify the limitation of IVR, the identification of the border is inevitable. We remind that asymptotic (i.e., semiclassical) analysis has an ability to treat exponentially small quantities. This suggests some asymptotic approach may reveal the limitation of IVRs with much better accuracy.

In this paper, we examine an evaluation of a tunneling tail, by the Herman-Kluk formula, with asymptotic analysis. Here we focus on the Feynman kernel, rather than energy spectra or correlation functions. This facilitates to identify the origin of discrepancies.

We here examine a single degree of freedom system that is described by a Hamiltonian $H = -gp^3/3$, where q and p are the position and the momentum of the system, respectively. We assume that the strength of folding g is positive. This is a canonical model that describes a nonlinear folding process in classical phase space (see, Fig. 1 (a)) [3]. When both the initial and the final states of Feynman kernel are eigenstates of the position operator, the nonlinear folding dynamics produces a caustic. Note that in generic, nonlinear systems induce foldings in general. Dynamics locally around each foldings are described by the canonical Hamiltonian with appropriate rotations and scaling in phase space (see, e.g., Fig. 1 (b)).

The Feynman kernel of a time evolution of interval $[0, \tau]$ $(\tau > 0)$ in the position representation $K(q) \equiv \langle q| \exp(-iH\tau/\hbar)|q = 0\rangle$ is expressed exactly with Airy function

$$K(q) = A_i(q/l)/l \tag{1}$$

where $l \equiv (\hbar^2 g\tau)^{1/3}$ is a characteristic length for a penetration into the classically forbidden region $q > 0$. The asymptotic evaluation of the integral representation of the Feynman kernel

$$K(q) = \int dp \langle q|p\rangle e^{+igp^3\tau/(3\hbar)} \langle p|q = 0\rangle \tag{2}$$

gives a leading semiclassical approximation $K_{\rm SC}$, which are composed by classical trajectories. On one hand, in the classically allowed region $q < 0$, we have a superposition of incoming and outgoing waves:

$$K_{\rm SC}(q) = \frac{1}{\sqrt{\pi} l (|q|/l)^{1/4}} \cos\left\{ \frac{2}{3}\left(\frac{|q|}{l}\right)^{3/2} - \frac{\pi}{4} \right\}. \tag{3}$$

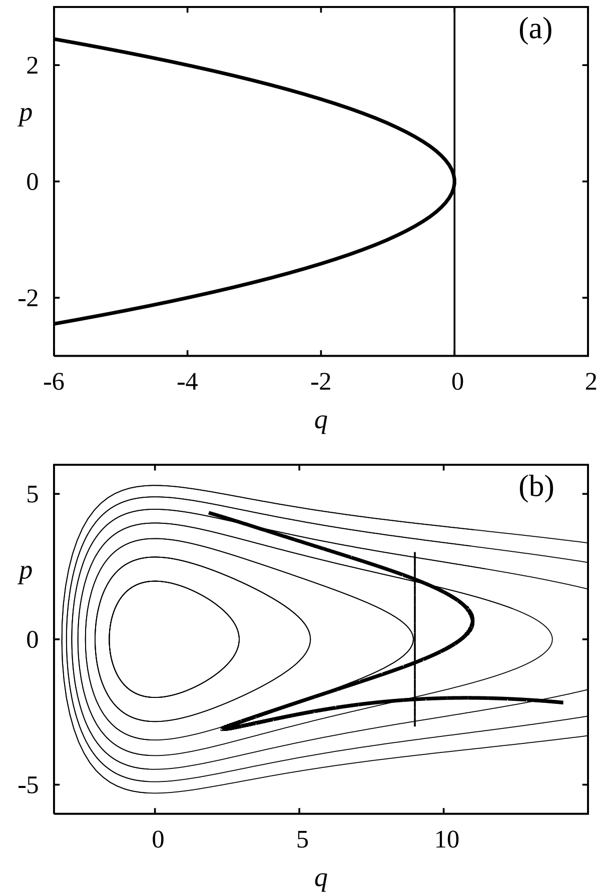

FIG. 1: Evolutions of classical manifolds in phase space. (a) With the folding Hamiltonian $H = -gp^3/3$ [3]. The initial manifold (thin line) is at $q = 0$. After a time interval τ, the manifold folds (thick line). The caustic in the position representation is at $q = 0$. (b) With a nonlinear oscillator $H = \frac{1}{2}p^2 + V(q)$, where $V(q) = D\{(1 - e^{-\lambda q})^2 - 1\} + \frac{1}{2}(1-\epsilon)q^2$, $\epsilon = 0.975$, $\lambda = 1/\sqrt{12}$, and $D = \epsilon/(2\lambda^2)$ (Contour lines of $V(q)$ are thin) [15]. The initial manifold $\{(q,p)|q = 9, -3 < p < 3\}$ (vertical line) mimics an eigenstate of the position operator, with an energy cutoff. The corresponding final manifold (thick line), at $t = 18$, has two prominent caustics in the position representation .

The corresponding classical trajectories are characterized by their momentum, which are conserved quantities: $p_\pm(q) = \pm\sqrt{|q|/(\tau g)}$. On the other hand, in the classically forbidden region $q > 0$, we have a tunneling tail:

$$K_{\mathrm{SC}}(q) = \frac{1}{2\sqrt{\pi} l (q/l)^{1/4}} \exp\left\{-\frac{2}{3}\left(\frac{q}{l}\right)^{3/2}\right\}. \tag{4}$$

The momentum of the tunneling trajectory is pure imaginary: $p_0(q) = i\sqrt{q/(\tau g)}$. Between

these regions, at $q = 0$, these classical trajectories merge to produce a caustic. The change of asymptotic expansions for different signs of q is controlled by Stokes phenomenon [16].

In the following, the corresponding Herman-Kluk kernel [10] is examined:

$$\begin{aligned} K^{\mathrm{HK}}(q) &= \int\int \frac{dq_0 dp_0}{2\pi\hbar} \langle q|\varphi^\gamma(q_\tau, p_\tau)\rangle C(q_0, p_0, \tau) \\ &\quad \times e^{iS_\tau(q_0,p_0)/\hbar} \langle \varphi^\gamma(q_0, p_0)|q = 0\rangle, \end{aligned} \tag{5}$$

where γ (> 0) determines the width of Gaussian packet $\langle q|\varphi^\gamma(q_0, p_0)\rangle = (2\gamma/\pi)^{1/4} \exp\{-\gamma(q - q_0)^2 + ip_0(q - q_0)/\hbar\}$, (q_τ, p_τ) is the classical trajectory at time $t = \tau$, emitted from (q_0, p_0) at $t = 0$, $S_\tau(q_0, p_0)$ is the classical action along the time evolution, and $C(q_0, p_0, \tau) = \{\partial q_\tau/\partial q_0 + \partial p_\tau/\partial p_0 - 2i\hbar\gamma\partial q_\tau/\partial p_0 - (2i\hbar\gamma)^{-1}\partial p_\tau/\partial q_0\}^{1/2}/\sqrt{2}$. For the folding Hamiltonian, $C(q_0, p_0, \tau)$ have a branch point at $p_{\mathrm{I}} = i/(2\hbar\gamma\tau g)$. After the integration of the variable q_0, $K^{\mathrm{HK}}(q)$ (5) becomes

$$K^{\mathrm{HK}}(q) = \int \frac{dp}{2\pi\hbar} C(p, \tau) e^{-\phi_\tau(p)}, \tag{6}$$

where $C(p, \tau) = (1 - p/p_{\mathrm{I}})^{1/2}$ and $\phi_\tau(p) = \gamma(q + \tau g p^2)^2/2 - ipq/\hbar - i\tau g p^3/(3\hbar)$. The integral (6) has three saddle points $p = \pm\sqrt{-q/(\tau g)}$ and p_{I}. The former momenta $p = \pm\sqrt{-q/(\tau g)}$ correspond to the classical momenta $p_\pm(q)$ in the classically allowed region and $p_0(q)$ in the classically forbidden region. The latter momentum p_{I}, which is the branch point of $C(p, \tau)$, appears only in the semiclassical analysis of $K^{\mathrm{HK}}(q)$. Note that all the saddle points need not to make contributions to the semiclassical kernel, due to the Stokes phenomena. Actually, in the classically allowed region $q > 0$, there are the contributions only from $p = p_\pm(q)$ (FIG. 2(a)). Hence the leading semiclassical evaluation of $K^{\mathrm{HK}}(q)$ agree with $K_{\mathrm{SC}}(q)$. Thus the asymptotic equivalence between Herman-Kluk kernel and quantum theory holds for $q < 0$ [6].

In the semiclassical evaluation of $K^{\mathrm{HK}}(q)$ in the classically forbidden region $q > 0$, there is a length scale $l_\gamma = \gamma^{-2} l^{-3}/4$ that divides the tunneling tail into two regions: a "shallow" region $0 < q < l_\gamma$ and a "deep" region $q > l_\gamma$. In the shallow region, the semiclassical evaluation of $K^{\mathrm{HK}}(q)$ has only a single contribution from the classical trajectory $p = p_0(q)$ (FIG. 2(b)). Hence the asymptotic equivalence between Herman-Kluk kernel and quantum theory holds both classically allowed region and the shallow tunneling region $q < l_\gamma$. This is a promising evidence that collections of real classical trajectories can describe *shallow* tunneling tails, through IVR techniques. This is the first example to show that IVR technique can describe a classically forbidden process with an analytical argument.

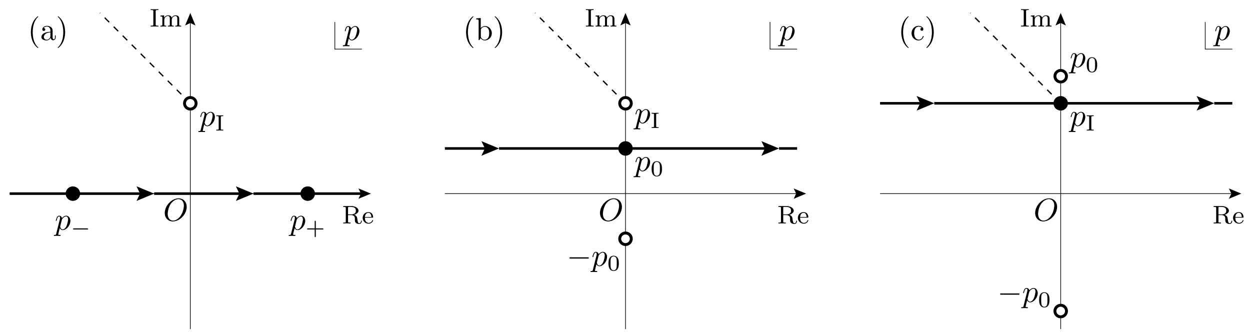

FIG. 2: Locations of saddle points (closed and open circles) and integration paths (thick lines) for the asymptotic evaluations of the integral (6), for (a) the classical region $q < 0$, (b) the shallow tunneling region $0 < q < l_\gamma$, and, (c) the deep tunneling region $q > l_\gamma$. Dashed lines emanating from the branch point p_I are branch cuts. Closed and open circles are contributing and non-contributing saddle points, respectively.

On the other hand, in the "deep" tunneling region, we found a discrepancy: Due to Stokes phenomenon, the contribution from "conventional" tunneling trajectory $p = p_0(q)$ disappears, and in turn, the contribution from the classical trajectory $p = p_\mathrm{I}$, which is an artifact of the Herman-Kluk kernel, appears (FIG. 2(c)). The resultant semiclassical evaluation of $K^{\mathrm{HK}}(q)$ is

$$\begin{aligned} K^{\mathrm{HK}}_{\mathrm{SC}}(q) \;&=\; \frac{\Gamma(3/4)}{2\pi l(\gamma l^2)^{1/4}} \left(\frac{l}{q - l_\gamma}\right)^{3/4} \\ &\quad\times \exp\left\{-\frac{\gamma}{2}(q + l_\gamma)^2 - \frac{4}{3} l_\gamma^2\right\}. \end{aligned} \tag{7}$$

Note that $q = l_\gamma$ is a caustic, which is an reminiscent of the Stokes phenomena. At the same time, the asymptotic form of tunneling tail $\sim \exp(-\gamma q^2/2)$ for $q \gg l_\gamma$, which sensitively depends on γ, is qualitatively different from $K_{\mathrm{SC}}(q) \sim \exp\{-2(q/l)^{3/2}/3\}$ (4). Thus the breakdown of the asymptotic equivalence is evident.

In the argument above, the discrepancy between Herman-Kluk kernel and Feynman kernel comes from (1) a nonlinear folding dynamics in the corresponding classical phase space, and (2) the appearance of artificial tunneling trajectory. When the folding dynamics is not significant (this is the case before Ehrenfest time), there is a workaround to remove the contribution from the artificial tunneling trajectories, by adjusting the value of γ in the Herman-Kluk kernel to push p_I deeper in the complex plane (see., Fig. 2). Indeed, this is a known strategy, which is proposed by Kay, to reduce the magnitude of the error of Herman-

Kluk method, and the strategy succeeds to a certain extent [14, 17]. However, in generic, nonlinear systems, it will become difficult to carry out such workarounds in practice, due to the emergence of multiple foldings (see, e.g., Fig. 1 (b)).

We summarize this paper. With an exactly solvable model that describes nonlinear folding process in corresponding classical phase-space dynamics, we identified a boundary between a shallow and deep tunneling regions for the Herman-Kluk kernel. In the former region, the Herman-Kluk kernel and quantum theory are asymptotically equivalent. Hence there remains a hope that Herman-Kluk kernel describes classically forbidden process to a certain extent. However, in the deep region, the breakdown of the asymptotic equivalent is shown. Besides Herman-Kluk method, other IVR approaches, in particular, which are based on Kay's framework [6], will have similar scenario on successes and failures in descriptions of classically forbidden phenomena. We expect that the nonlinear folding Hamiltonian employed here is a good test-case not only for the Herman-Kluk method, but also for various semiclassical method. We remind that this model was employed to elucidate the limitation of single trajectory approximation of semiclassical coherent-state path integrals [3, 18]. At the same time, it is highly desirable to develop a convenient way to find the boundary between the shallow and the deep regions of Herman-Kluk method, though the present analysis requires a full knowledge of the Stokes phenomena to determine the boundary.

Acknowledgments

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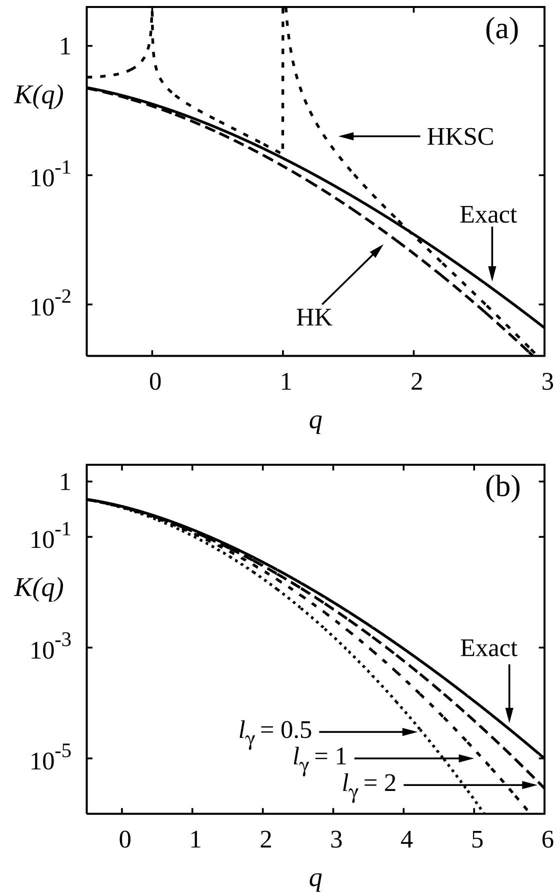

FIG. 3: Comparisons of exact, Herman-Kluk (HK), and semiclassical Herman-Kluk (HKSC) evaluations of the Feynman kernel $K(q)$, in particular, its tunneling tail $q > 0$. The penetration depth is $l = 1$. (a) With $l_\gamma = 1$, all three theories are shown. At the (conventional) turning point $q = 0$, the semiclassical Herman-Kluk encounters caustic. In the shallow region $0 < q < l_\gamma$, the discrepancy between quantum theory and Herman-Kluk method is not significant. On the other hand, at $q = l_\gamma$, the semiclassical Herman-Kluk encounters another caustic. Note that the conventional semiclassical theory meets the caustic at $q = 0$, the conventional turning point, only. In the deep region $q > l_\gamma$, the tunneling tails of quantum theory and Herman-Kluk formula take qualitatively different shape (see, Eq. (4) and Eq. (7)). (b) Comparison of quantum theory and Herman-Kluk formula, with several values of l_γ. In order to reproduce the shape of deep tunneling tail by Herman-Kluk method, we need larger l_γ [14].